ISENTROPIC COMPRESSION OF ARGON

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Introduction

We are studying the transition of argon from an insulator to a conductor by compressing the frozen gas isentropically to pressures at which neighboring atomic orbitals overlap sufficiently to allow some electron motion between atoms. Argon and the other rare gases have closed electron shells and therefore remain monatomic, even when they solidify. Their simple structure makes it likely that any measured change in conductivity is due to changes in the atomic structure, not in molecular configuration. As the crystal is compressed the band gap closes, allowing increased conductivity. We have begun research to determine the conductivity at high pressures, and it is our intention to determine the compression at which the crystal becomes a metal. Details of the equation-of-state calculations will be given by Kress and Collins, paper P2-1 of this conference. Here we describe the experimental techniques.

To make the compression as easy as possible (and to simplify the equation-of-state calculations) it is important to keep the sample from heating. Ideally the compression would occur slowly, for example in a diamond anvil cell. However that method cannot attain the pressures of 500 to 700 GPa, or higher, estimated to be necessary for metalization of argon, and the samples would be too small to allow simple, direct methods of measuring their bulk properties, such as the conductivity. Shock compression using explosives or gas guns is often used to get to the pressure range we need, but shock heating greatly reduces the compression. We have chosen instead to do the compression isentropically in an MC-1 flux compression generator[1].

Experimental Set-up

A schematic of the experiment is shown in Fig. 1. Frozen argon gas is contained in a conducting tube in the center of the MC-1. A probe on the axis measures the sample conductivity to the tube wall. A capacitor bank discharge introduces a solenoid field of 14 T outside the tube, the explosive is detonated to compress the magnetic field volume, and the field increases to around 500 T in a matter of microseconds, squeezing the tube and compressing the sample inside. As long as care is taken to avoid allowing the imploding MC-1 to strike the tube, the compression is essentially isentropic. When the tube reaches its minimum volume a radiograph is taken to determine the inner radius and the compression.

The MC-1 generator, which has been described in detail in the literature[1], will be outlined briefly here. On the outside is a cylindrical explosive charge, 30 cm diam by 18 cm long, that is detonated simultaneously around its outer surface, providing a cylindrical implosion of the material inside. Lining the inside of the explosive is a cylinder about 15 cm in diam, 30 cm long, and 6 mm thick, called the first, or outer, cascade. It is fabricated as an epoxy matrix containing several thousand closely-packed, insulated copper wires wound in a two-turn solenoid-coil configuration. A second similar cascade rests coaxially inside. It is thinner and has its wires parallel to the axis, and it has a radius of about 3 cm. Inside the second cascade goes the experimental sample. Current from an external capacitor bank discharge flows through the first cascade to provide a seed magnetic field.

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14. ABSTRACT

We are studying the transition of argon from an insulator to a conductor by compressing the frozen gas isentropically to pressures at which neighboring atomic orbitals overlap sufficiently to allow some electron motion between atoms. Argon and the other rare gases have closed electron shells and therefore remain monatomic, even when they solidifi. Their simple structure makes it likely that any measured change in conductivity is due to changes in the atomic structure, not in molecular configuration. As the crystal is compressed the band gap closes, allowing increased conductivity. We have begun research to determine the conductivity at high pressures, and it is our intention to determine the compression at which the crystal becomes a metal. Details of the equation-of-state calculations will be given by Kress and Collins, paper P2-1 of this conference. Here we describe the experimental techniques.

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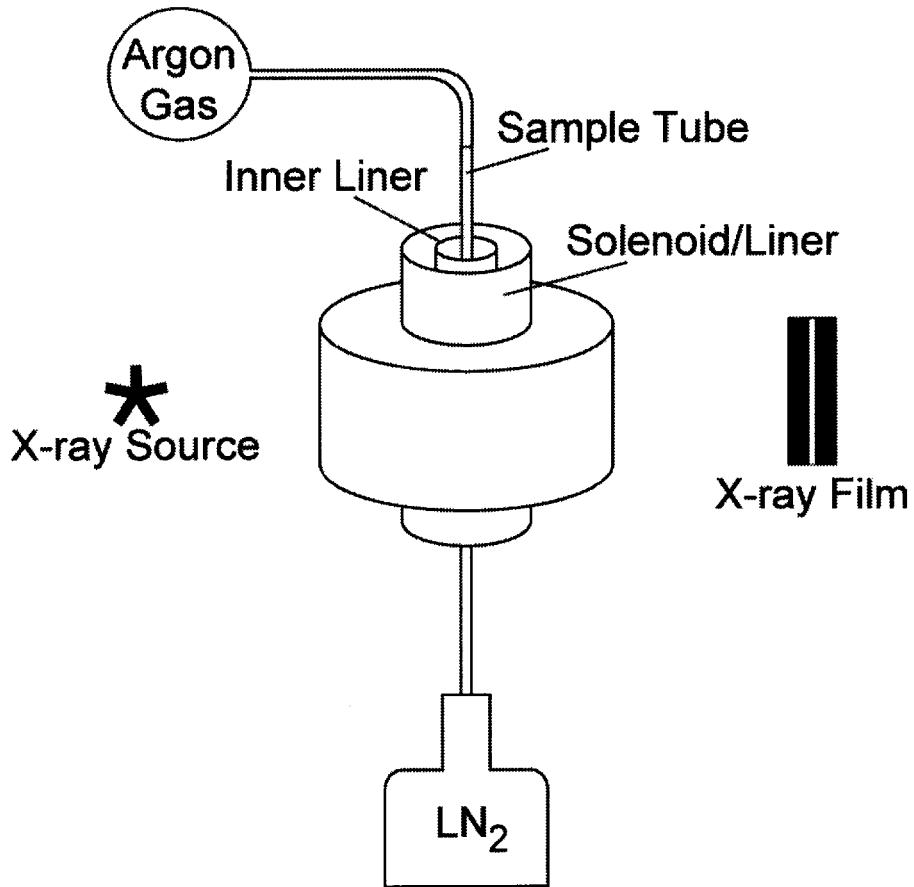
Figure 1. Schematic of the isentropic compression experiment.

At peak current time the detonating explosive front reaches the cascade and shocks it sufficiently to turn it into a conductor and to drive it radially in with a velocity of 4 km/s. The flux trapped within the imploding cascade when it becomes conducting is compressed. Since the unshocked cascade configuration does not support eddy currents, the flux can diffuse through the second cascade to fill the volume outside of the sample tube. The second cascade is sized so that it is struck by the first cascade about when it begins to break up from instabilities. When struck, the second cascade also becomes a conductor and is driven inward with most of the flux trapped inside its walls. The sample tube is made a little smaller than the inner cascade. The flux between them compresses the sample and keeps the cascade from striking it and causing a shock.

Figure 2 shows a schematic of the sample. The argon is contained in a 2-mm-thick copper tube with a 1-mm-thick tungsten layer on the inside wall to improve the radiograph contrast. Its inside diameter is 14 mm, and it is longer than the 18-mm length of the explosive, so that there is a transition region from the uniformly-imploded center to the non-imploded ends. One end is made of a Teflon cone, and the other is a copper wall with a Teflon washer inside. Both ends are outside the imploded region. The tube is filled with argon, which is frozen by immersing the bottom end of the tube in liquid nitrogen until just before the shot is fired. A careful study was made to ensure that the filling technique does not leave voids in the argon.

180

160

Ar

90

Axial Electrode

Compression Tube

W 13.8 x 15.8
Cu 15.8 x 19.8

Teflon

R

Measuring Circuit

V   I   R   r

$$r = \frac{R}{\dfrac{v_0}{v} - 1}$$
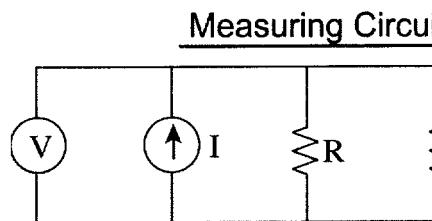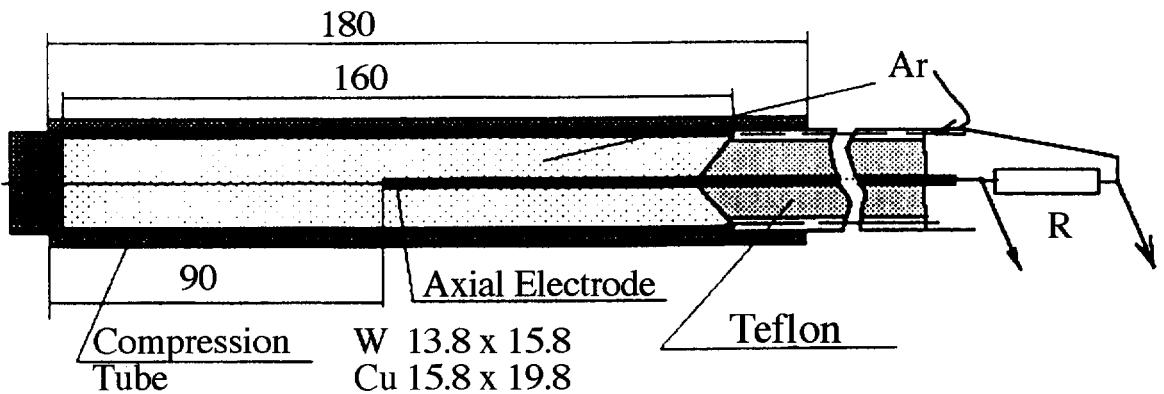
R = reference resistor
r = Argon resistance

Solving Poisson equation gives capacitance, C. Then resistivity is

$$\rho = \frac{1}{\epsilon_0}\, rC$$

Figure 2. The sample tube, conductivity probe, and reference resistor.

The conductivity probe consists of a 2-mm-diam wire reaching part way into the sample from the end of the axis with the Teflon cone. All but the tip of the probe is insulated with a varnish coating, so when the argon becomes conducting, current flows from the probe tip to the walls of the tube. A reference resistor of about 1-Ω connects the probe to the tube beyond the Teflon end. A small capacitor is discharged, sending current in parallel through the argon and the resistor. We can determine the argon resistivity knowing the reference resistor, the ratio of the voltage drop across the resistor at the time of interest to that when there is no conductivity, and the probe geometry. Figure 3 shows a measurement of the voltage drop across the reference resistor as it varied with time on a 1995 shot. There is a slow RC decay of the capacitor circuit, a sharp dip at compression time (51 μs), a return to the pre-compression voltage when the sample begins to decompress, and a drop to zero when the probe is shocked and destroyed.

The sample diameter is measured near minimum volume using a 60-MeV air-core betatron as a source of high-energy x-rays for a radiographic image. Fast x-ray film is placed in a steel vessel where it is protected from shrapnel and the explosive shock. The experiment is located between the source and film. By measuring the magnification of the optical system and the inner diameter of the tube on the transmission radiograph we can determine the sample compression at radiograph time. We cross-time the explosives detonation, the conductivity signal, and that from a photodiode measuring the betatron flux. The timing of the radiograph relative to the implosion is presently determined by calculations of the inner cascade position and assumes that the maximum compression and maximum conductivity coincide in time. For future experiments we will use an x-ray-to-visible light conversion system and a camera to record images at three times during the implosion to get the tube radius as a function of time. With this information we can normalize the
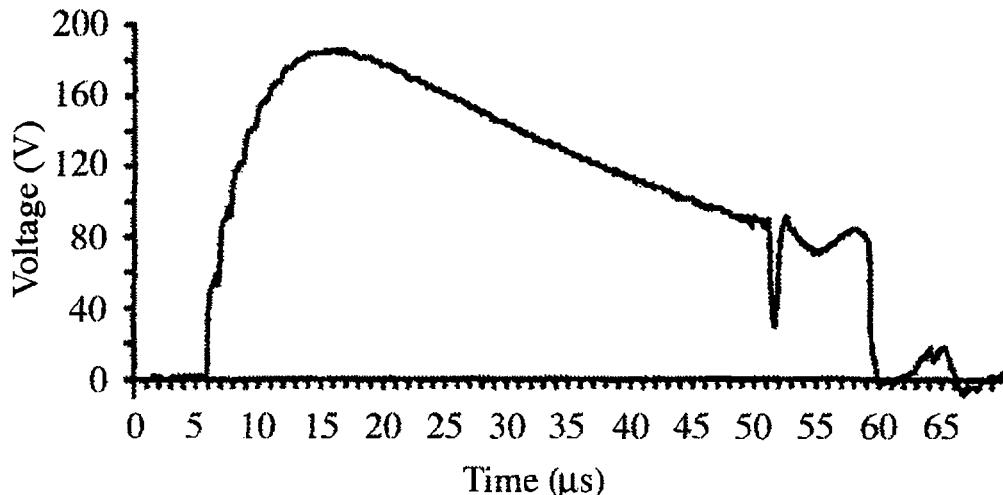
Figure 3. Reference resistor voltage v. time. At 50 μs the conductivity of the sample increased, causing a drop in the reference resistor current. Shortly after, the sample decompressed enough that the conductivity dropped again. At 60 μs the probe broke.

calculated value of r(t) to the measurement. The betatron has been modified to emit the proper pulse structure, and a feasibility experiment was fielded with a scintillator and streak camera.

Experimental Results to Date

Our resources allow us to make one short campaign in Russia each year to gather data using equipment that is ordinarily used on other experiments at Los Alamos. To summarize the work so far, we made a feasibility experiment in 1994, in 1995 we measured conductivity and learned that the argon did not metalize as expected, and in 1996 we increased the compression, decreased the probe sensitivity, and began improvements of the experimental techniques.

1995 Experiments. From the data shown in Fig. 3, $V_{min} / V_0 = 0.3$, and from this ratio we determined a maximum conductivity of 8 ± 2 $(\Omega \, cm)^{-1}$ for a peak compression of 5.5 (the compression was 4.9 ± 0.2 at radiograph time) in the 1995 experiment. Calculations indicate a maximum pressure of $P \approx 6$ Mbar and a temperature of $T \approx 1500$ K. Typical metals have a conductivity of thousands per Ω-cm, and for argon the expected conductivity was around 10^3 $(\Omega \, cm)^{-1}$. Apparently at this compression the electrons can make indirect transitions from atom to atom but cannot yet freely move around as in a metal.

The radiograph for the 1995 shot showed that the compression was uniform along the axis and that there was sufficient space between the inner cascade and the tube to be sure that there was no shock. Experiments the following year verified that the reference resistor was sufficiently far from the transition between the imploded and non-imploded regions that it was not damaged in the implosion.

1996 Experiments. For the 1996 series we set several goals. First, we lowered the seed magnetic field to around 10 T to allow faster motion of the cascades and a higher final pressure. This carries

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some risk because there is more danger of a collision of the inner cascade with the tube, causing a shock. Calculations show there is still no collision, but it is difficult to confirm this from the radiographs. Furthermore the faster implosion has a pressure peak that is sharper in time, making it more difficult to know how close to peak compression the radiograph was taken. Second, we modified the conductivity probe geometry to decrease its sensitivity. (The reference resistor cannot be made significantly smaller because of the contact resistance.) Finally, we used the betatron multi-pulse structure, and we set up a 3-cm-diam NaI scintillator just behind the film pack, along with optics to transport the light to a streak camera, to image the transmitted x-ray beam. The 1996 series consisted of five shots. The first was to test the lower seed field, the new conductivity geometry, and the three-pulse imaging system. The second was a repeat, but without the multi-pulse imaging. The final three shots were to examine the probe insulation and to study ways to improve it to decrease the probe sensitivity.

The sensitivity of the conductivity probe was decreased by reducing the diameter of the center conductor to 0.4 mm and by lining the inner wall of the tube with a 0.3-mm-thick layer of Teflon, except for a 5-mm-long ring about 50 mm axially from the probe tip. The ring contained a 0.3-mm-thick layer of aluminum to allow a small conducting path through the Teflon to the tube wall. The center conductor was also coated with 0.3 mm of Teflon in addition to the varnish, again leaving the probe tip uninsulated, and the tip was rounded into a hemisphere to simplify the geometry calculations a bit. Although the insulation properties of Teflon had been tested to above 4 Mbar, we found that apparently it breaks down at these higher pressures, > 7 Mbar. As a result the full surface of the tube was able to conduct current, as if the Teflon were not present, giving a signal similar to the 1995 experiment. Notably absent on the first two 1996 shots was any significant conductivity increase resulting from the increased pressure, and the conductivity appeared at a time very similar to the 1995 experiment.

To provide more information on the tube radius, r(t), we configured the betatron to provide three pulses with intervals of about 2.0 and 0.5 μs between the first, second, and third pulses. They were timed so that the third pulse would arrive near maximum compression time. The total number of electrons in the three pulses is fixed by the betatron current, so we arranged to have the pulse amplitudes increase sequentially to compensate for the decrease in transmission through the generator as the compression increases. In addition the amplitude of the third pulse was doubled to allow for a readable film record at maximum compression time without excessive background from the early pulses. The width of the third pulse was about 40 ns; the pulser broadened the first two pulses a little relative to the third. The betatron focal spot is elliptical, about 4 mm high by 1 mm wide, and it strikes a 2-mm-thick tungsten target to produce bremsstrahlung x-rays.

The recording was done in a shock-hardened containment vessel, called the "pig." The x-rays that were transmitted through the sample passed first through the x-ray film and then struck a 10-mm-thick NaI(Tl) scintillator. An optical relay system took the scintillator output light to the slit of a streak camera, also within the pig, but below the x-ray beam to reduce backgrounds in the image. The camera was rotated so that the slit was in the horizontal plane with time dispersed vertically. In this configuration we looked at a thin slice across the middle of the tube, and the camera streak record had three separate images defined by the betatron pulse structure. The optical resolution was limited to 2 line-pairs/mm, mainly by the scintillator, which was thicker than we would have liked because of the shortage of signal light. In 1996 we fielded the multi-pulse recording system on just one shot. We were able to see the edges of the imploding tube on the third pulse, but the first two pulse images were too faint to return useful data. The recording system is presently being redesigned and upgraded to increase the signal intensity and resolution.

The final three shots were made to understand where the insulator fails, to enable us to design a better probe for future shots. In shot 3 the Teflon insulator lined the entire length of the outer electrode. In shot 4 the center electrode was extended the full length of the argon sample and was insulated with Teflon along its whole length, while the outer electrode was left completely

uninsulated. These two shots gave conductivity data similar to the preceding shots, indicating that the Teflon apparently has no significant effect in this configuration, either at the inner or the outer electrode. Our conjecture is that during the implosion the outer Teflon sleeve stretches and tears near the ends of the compressed region, in the transition between the compressed and uncompressed tube wall, and the inner Teflon is perhaps destroyed by jets of material, probably also in or near the transition regions. Finally the fifth 1996 shot was made with the center probe shortened so that it did not enter the compressed region. This time the signal drop appeared nearly 2 μs later, about the expected time of probe destruction.

Conclusions

Since we observe conductivity signals in all of the experiments (except when the probe is removed) we know that both the Teflon and the argon are becoming conductive. However, we are not able to determine the conductivity because without knowing the Teflon insulation properties we cannot determine the geometric factor. We can bound it, however, as being $\sigma = 1/\rho \leq 8 \ (\Omega\,\text{cm})^{-1}$. because when the Teflon breaks down the dielectric constant is reduced (see equation in Fig. 2.)

The Teflon insulator problem remains perplexing, and we are now searching for other ways to better insulate the probe or to measure the conductivity without a probe.

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